

# PATENT SPECIFICATION

(11) 1303810

B7

1303810

## NO DRAWINGS

(21) Application No. 22650/69 (22) Filed 2 May 1969  
 (23) Complete Specification filed 1 May 1970  
 (44) Complete Specification published 24 Jan. 1973  
 (51) International Classification C11D 11/00  
 (52) Index at acceptance  
 C5D 6A10 6A5C 6A5F 6A9 6B11C 6B11D 6B12B1 6B12F2  
 6B12G2A 6B12G2B 6B12G3 6B12G6 6B12K2  
 6B12L 6B12M 6B12N2 6B7 6B8 6C8 6D  
 (72) Inventors DONALD ARCHIE HUDSON and  
 WILLIAM ARTHUR SENIOR

(19)



## (54) POURABLE LIQUID COMPOSITIONS

(71) We, UNILEVER LIMITED, a company registered under the laws of Great Britain, of Port Sunlight, Wirral, Cheshire, England, do hereby declare the invention for 5 which we pray that a patent may be granted to us and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to pourable 10 liquid compositions for cleaning or rinsing operations.

Accordingly the compositions of the present 15 invention comprise a clear, liquid medium and a visually distinct component of particle size at least 0.5 mm diameter, stably dispersed and stably suspended in the medium.

The component may be in solid or liquid form.

By the "medium" is meant the continuous 20 phase of a multi-phase system.

By "stably" dispersed is means that the dispersed component remains discrete, i.e. it does not coalesce, flocculate or agglomerate over a period of at least six months at 25°C.

By "stably" suspended is meant that the component remains suspended in the medium for at least six months when stored at 25°C and does not sediment when the composition is disturbed in transportation, storage and 30 use.

By "visually distinct" is means that when the composition is seen by an average human observer under conditions of illumination by normal daylight, artificial light or in the 35 case of systems containing fluorescent material, ultra violet light, the dispersed component can be recognised as discrete entities when viewed at a distance of between 10 inches and 40 inches.

These compositions have the advantages 40 known in other industrial fields that the visual distinctness apparent in them can be associated in the mind of the purchaser/user with, for example, the origin of the compositions, a 45 performance attribute or a product quality.

The present compositions have additional

practical advantages in the context of immiscible system accruing from their stable suspension attribute. They provide pre-determined proportioning of the medium/component without need for shaking by the user before dispensing, and each portion dispensed will necessarily include the correct proportions of medium and component. This is particularly helpful in compositions for use in hair washing. This advantage is even more important in the compositions of the invention which have several components. The dispensing of, for example, multi-ingredient cleaning compositions which are not in the stable suspension and stable dispersion form of the compositions of the present invention, requires either complex metering devices, agitation to produce a temporary dispersion or production of the composition initially as a micro dispersion if correct proportioning of the ingredients is to be achieved. The first of these techniques is expensive, the second is inconvenient and the third method gives rise to an opaque product which may not be commercially desirable.

Another advantage of the present compositions is that the component, because it is in visually distinct form, can be used as a tracer for measuring the exit flow rate of a pourable composition, i.e. dosage can be controlled easily by the user simply by visual checking of the component as the composition is poured from a container.

The present invention is concerned with compositions in which the medium is clear. Purchasers/users of liquid cleaning compositions frequently prefer transparent products. It is often both difficult and expensive to include certain necessary ingredients into liquid cleaning compositions whilst maintaining clarity of the product because of the need to use hydrotropes. Furthermore, hydrotrope inclusion in a suspending liquid can often impair suspending properties. Thus a composition in which the dispersed component includes any material which would otherwise

50

55

60

65

70

75

80

85

90

render the medium opaque is commercially valuable. There is commercial value also in locating in the dispersed component any agent which detracts from the colour of the composition, for example a light-unstable germicide such as tribromosalicylanilide or a dark coloured raw material, for example coconut monochanolamide. Similarly, agents which detract from the consumer acceptability, e.g. 5 objectionable smell can also be located in the component. 70

Accordingly, in the present invention, the medium is transparent, i.e. it has a turbidity of five or less, preferably less than three. The 10 "turbidity" of a system is a measure of the amount of light which the system scatters. If a system, either liquid or solid, is placed in a visible spectrophotometer an optical density of the system at any given wavelength 15 of light can be measured according to standard procedures. The extinction of the system is  $2.303 \times$  the optical density as measured in a 1 cm cell. The turbidity of the system is that part of the extinction which is not due to 20 absorption, i.e. is entirely due to scattering. Turbidity is defined by H. C. van de Hulst "Light scattering by small particles", John Wiley, 1957. In the preferred compositions 25 the amount of dispersed component is not more than 10% by volume of the total composition. The component may be in any convenient form, for example spheres of 4 mm diameter at a concentration of 1 sphere per  $\text{cm}^3$  of medium (3.35%, by vol.) or by spheres of 1 mm diameter at a concentration of 10 spheres 30 per  $\text{cm}^3$  of medium (0.52% by vol.). 35

Alternatively, the concentration of dispersed component may be from 20—80% by volume of the total composition. 40

The media may be aqueous, non-aqueous, a mixture of miscible liquids or a solution of one or more solid materials in an aqueous, or non aqueous liquid or in a mixture of miscible liquids. The medium may be 45 useful for some purpose additional to that of simply acting as a vehicle for the suspended component or components. The media may be coloured or water-white subject to the provisos to be discussed later. It should also, 50 of course, in the case of cleaning or rinsing compositions for personal use be non-toxic and non-irritant to the skin. The medium must be capable of stably suspending the component and be inert to the exterior of the 55 suspended component, preferably the medium will have rheological properties which are non-Newtonian.

Ideally these rheological properties should 60 correspond to those of a so-called Bingham body. By virtue of its internal structure such a medium will exhibit a yield value from which it is possible to calculate the maximum size of particle which can be stably suspended for a given difference in density 65 between the medium and dispersed component.

For example, the yield value required by the medium to suspend spherical globules of any given density is given by the formula: Yield value (dynes per square cm) is greater or equal to  $654 \frac{r}{\Delta d}$  where  $r$  is the radius of the globule in cms and  $\Delta d$  is the difference in densities (gms per  $\text{cm}^3$ ) between the two phases irrespective of which is denser. This formula applies for spherical homogeneous globules; analogous formulae can be derived for globules of more complex shapes and internal structures. Preferably the yield value should be as low as possible in order to facilitate pourability of the composition. Thus a medium having a yield value of  $2 \text{ dynes cm}^{-2}$  is readily pourable and will suspend spheres of up to 2 mm diameter, providing that the density difference does not exceed 0.03 gms per  $\text{cm}^3$ . The yield value of the medium can be determined with the aid of a low shear rate viscometer as described by van den Tempel (J. Phys. Chem. 67 731 (1963) using the method of Green "Industrial Rheology and Rheological Structure", John Wiley 1949 p. 54. 75

Some selected media will possess the required rheological properties by virtue of their inherent character. 80

Other media will require addition thereof of a suspending agent, such as a swelling clay, a polymer gelling agent, a soluble gum, or a colloidal or pyrogenised silica, in order that the selected component may be stably suspended whilst remaining pourable. The suspending agent, if used, should be selected so as not to interfere with the desired optical properties of the liquid medium nor should it be chemically incompatible with the medium or the dispersed component. 85

Specifically suitable suspending agents, 90 amongst the many commercially available, for inclusion in a medium when that medium has not the correct rheological properties, are Carbopol (RTM B.F. Goodrich Chemical Company — a carboxy vinyl polymer of extremely high molecular weight), Laponite S (RTM—a synthetic swelling clay from Laporte Industries Limited), Benagel EW (RTM—a montmorillonite clay from Berk Limited), Cabosil (RTM—a pyrogenised silica from Cabot Chemical Corporation) and a xanthan gum. The amount and nature of the suspending agent added will depend on the 100 rheological properties required, the nature of the suspended component (density size etc.) and the nature of the medium in which the component is to be suspended. Also the 105 conditions of use of the composition e.g. pourability and conditions for storage must be considered. 110

It is also desirable that the medium should exhibit a minimum of thixotropy, i.e. when the internal structure is disrupted by the 115 stresses involved in pouring, recovery of the structure upon removal of the stress should 120

125

130

occur within a few seconds. This characteristic will effectively eliminate any separation of the dispersed components which could occur during the period required for recovery of the internal structure.

The amount of suspending agent will generally be not more than 5% by weight of the medium but may, in rare cases, be somewhat more.

The component to be stably suspended by the medium must be visually distinct therefrom. The visual distinctness can be obtained by way of selection of shape of the component, for example by spherical elongated or pear-shaped globules, provided that the size and also the optical characteristics to be discussed below are such as to render them visually distinct. The globules which can be solid or liquid can have a geometrical shape and should have linear dimensions of greater than 0.5 mm but should not be so large as to overcome the ability of the medium to suspend them. The upper limit of the component dimension is determined by such factors as the form of the pack to be used for marketing of the composition and the pourability character required. This will usually be not more than 1 cm in diameter. The preferred range of particle size of the component, being between 1 mm and 1 cm in diameter.

The visual distinctness can be achieved by manipulation of any of the following conditions:

- (a) The geometry of the component;
- (b) The optical constants of the medium and the component, viz. the refractive indices, the absorption co-efficients, and the emission co-efficients; (the overall appearance of the compositions may of course be modified by the choice of the light source used for viewing).
- (c) The distribution of the component in the medium e.g. evenly, as in compositions approaching a close packed configuration or randomly as in compositions having an appreciably less dense packing.

The present invention is also concerned with compositions for cleaning and rinsing operations which include more than one visually distinct, stably suspended, stably dispersed component.

As the dispersed component is preferably responsible for a technically advantageous property it must be responsive to the in-use environment so to release the agent which provides the advantage during use of the compositions.

The compositions of the present invention are for use in cleaning or rinsing applications, particularly in the cleaning of surfaces when the compositions will necessarily include at least 1% of a cleaning agent, viz. an agent or agents selected from inorganic compounds such as acids e.g. phosphoric acid and acid salts such as sodium dihydrogen phosphate and alkalis, e.g. alkali metal phosphates, tri-

sodium orthophosphate, silicates, and carbonates, and organic compounds such as soap e.g. derived from  $C_6$ — $C_{18}$  fatty acids, natural or synthetic and synthetic detergents e.g. alkyl sulphates and alkyl aryl sulphonates. The compositions for the cleaning operation must be capable of effecting cleaning as opposed to mere wetting. In the preferred cleaning compositions of this invention the media have cleaning properties. They may be the aqueous or non-aqueous liquid light, medium or heavy duty cleaning formulations. There are many of these commercially available and well known in the literature as, for example, the generally accepted typical formulation given below. A typical dishwashing formulation consists essentially of 15—20% alkyl benzene sulphonate, 13—20% alkyl ethoxylated sulphate and water; a typical fabric washing formulation consists essentially of 10—20% phosphate builder salt, 5—15% alkyl aryl sulphonate, some silicate and water; a typical hard surface cleaner consists essentially of 5—15% alkali metal pyrophosphate, a lather booster, a small amount of soap and a small amount of alkyl benzene sulphonate; a typical personal washing product consists essentially of a transparent physical mixture of mineral oil, paraffin waxes and emollient; a hair shampoo consists essentially of a synthetic sulphate surfactant, water and an ethanolamide of a higher fatty acid.

The foregoing cleaning media formulations are given as typical and other formulations well known in the art are suitable for the purpose of this invention.

The media for compositions for rinsing operations depends upon the specific operation in mind. Compositions within this invention are liquid fabric conditioners which are applied to washed fabric in a rinsing step subsequent to laundering and which can consist essentially of an aqueous solution of a cationic softener such as hexadecyl trimethyl ammonium bromide; rinses for washed fabric to impart crease-resistance, flame-proofing or protection against insecticidal attack; a machine dishwashing rinse aid such as an aqueous solution of a nonionic synthetic detergent typified by a Pluronic (RTM); a composition for personal use other than cleaning, e.g. a mouthwash or an emollient lotion for the relief of skin dryness.

The component to be stably suspended in either the cleaning or the rinsing compositions is preferably chosen not only for its visual distinctness but also for some advantageous technical property in the in-use context of the composition. The suspended component may provide the advantageous property directly, e.g. it may be a droplet of an agent which will provide the requisite technical effect such as a corrosion inhibitor in a metal cleaning composition or an emollient oil in a personal cleansing composition. The suspended com-

ponent may comprise a protective inert carrier around the agent which has the technical property, for example a liquid carrier such as an oil or a solid carrier such as a wax. The protective carrier isolates the agent from attack by the medium in which it is suspended.

In such "protective" compositions it is necessary if the advantageous property is to be obtained that the coating be releasable in the in-use context. Also that the protective carrier be inert to the medium and to the agent protected. Agents which may be used in "protected" compositions are bleaches, enzymes and perfumes.

The inclusion of agents which are not in thermodynamic equilibrium with the medium requires some sort of inert coating to physically separate the agents and the medium. Because the present invention is concerned with particles of at least 0.5 mm diameter, it has the advantage of reducing the amount of wall material compared with that required to enclose the same volume of agent in the form of smaller particles. This is economical and also reduces the amount of wall debris during use which can be disadvantageous in some applications.

The amount of technically useful material present in the suspended component may vary from 0.1 to 80% by weight of the total composition.

Release of the agent may be by virtue of solubility under the use conditions or abrasion or by way of response to change of temperature e.g. by melting.

A component may be introduced for its absorbency properties, i.e. a dehydrating agent may be stably suspended in a non-aqueous medium containing enzyme or other material sensitive to water. A component may be a free radical inhibitor included in media which are prone to light discolouration and odour deterioration.

In some cases, for example in multi-functional products, the medium itself can be a solvent for an agent in the dispersed component which provides the beneficial property.

The preparation of the pourable liquids of the present invention may be by simple dispersion of the selected particles into the selective medium whilst taking steps to avoid trapping any unwanted air bubbles and mechanical damage to the composition.

The invention will now be described by way of Examples.

55

### Example 1

A clear mild dishwashing liquid medium was prepared according to the following composition:

	%	60
Sodium N(2-hydroxyalkyl*)	10	
N-methyl taurate		
Sodium C <sub>11</sub> /C <sub>16</sub> alpha-olefin	15	
sulphonate		
Benzyl alcohol	5	65
Laponite S synthetic clay		
(5% dispersion)**	30	
Sodium chloride	2	
Water, perfume, dye, etc.	to 100	

\*A mixture of 75% C<sub>14</sub> and 25% C<sub>16</sub> straight hydrocarbon chains

\*\*Supplied by Laporte Limited, Luton, UK.

Rheological measurements on this liquid medium indicate that it has a yield point of 1 dyne cm<sup>-2</sup>, as determined by a viscometer capable of shearing the liquid at a rate of 10<sup>-3</sup> cm<sup>-1</sup>, and is readily pourable as evidenced by an apparent viscosity of 150 centipoise when measured at a shear rate of 1000 sec<sup>-1</sup>.

Droplets of lanolin, as a skin care aid were prepared by ultrasonically dispersing 10 parts of a liquid lanolin fraction and 1 part TiO<sub>2</sub> in a solution of 8 parts of gelatine (Isoelectric point, pH=4.9) in 82 parts of water. The dispersion at 30°C was stirred into a liquid paraffin oil phase held at 18°C; the droplets were filtered off, washed in ice cold detergent solution to remove entrained paraffin and washed with water. Subsequently they were immersed in 10% formalin for 5 minutes, rinsed, immersed in 25% sodium bisulphite for 15 minutes and then again rinsed thoroughly with water. The droplets have a gelling temperature of approximately 33°C.

The droplets, which were approximately 2 mm in diameter and of density 1.03, were dispersed in the dishwashing liquid at the rate of approximately 1 per cm<sup>3</sup>. They are readily dissolved within 1 minute when the dishwashing liquid containing the droplets is used for dishwashing with the wash solution at 40—45°C. The composition is stable for periods of up to 12 months at any temperature in the range 0—35°C.

70

75

80

85

90

95

100

**Example 2**

A clear dishwashing liquid medium was prepared according to the following composition:

5	Sodium alkylbenzene sulphonate*	%
		20
10	Sodium C <sub>14</sub> —C <sub>16</sub> alpha-olefin sulphonate	5
	Lauric monoethanolamide	5
	Sodium xylene sulphonate	7
	Ethyl alcohol	3
15	Laponite S synthetic clay (5% dispersion)	20
	EDTA	0.2
	Water, perfume, dye, etc.	to 100

\*Prepared from DOBS-83 alkylbenzene supplied by Shell Chemical Co. Ltd.

Droplets were prepared by first dispersing 20 6 parts of tribromosalicylanilide in a melt of 94 parts of paraffin wax (m.pt. 40°C). The liquid slurry was formed through a jet of 2.0 mm diameter at a column rate of 300 cm sec<sup>-1</sup> into a cooled container, the length of trajectory and temperature gradient being such that the liquid column broke into droplets approximately 4 mm in diameter which solidified before being collected at the base of the container.

25 30 The droplets were incorporated in the above dishwashing liquid to provide an average density of about 1 per cm<sup>3</sup>. They were suspended for up to a year and exhibited no discolouration during 3 months' storage in the

35 40 liquid in a clear container even under exposure to daylight. A control composition in which 0.2% tribromosalicylanilide had been directly incorporated in the same liquid exhibited marked discolouration during exposure to daylight.

**Example 3**

A clear rinse aid liquid for dishwashing machines was prepared from 1 part of Pluronic L61 (RTM), 0.25% Carbopol 940 (a carboxyvinyl polymer) (RTM) and 98.75 parts water and adjusted to pH 8.0 with sodium hydroxide solution. (Pluronic L61 is believed to be a block copolymer of polyoxypropylene and polyoxyethylene, 10% being the latter).

45 50 Glacial acetic acid was encapsulated in paraffin wax, m.pt. 55°C, by the biliquid column ejection method described in US patent 3,423,489. The operating conditions used to make the capsules were as follows:

60	Pressure applied to pot	10 psig
	Ejection velocity	4 m sec <sup>-1</sup>
	Temperature of wall material	100°C
	Temperature of core material	20°C
	Immersion depth	1.5 mm
	Orifice size	0.4 mm

This produced capsules approximately 1000  $\mu$  in diameter which contained about 50% by volume of acetic acid. They were incorporated in the rinse aid liquid at a level of approximately 50 capsules per cm<sup>3</sup>, so providing a 1% solution of acetic acid but without the disagreeable smell of this acid. In use, the hot rinsing water passed into the dishwashing machine, rapidly melted and dispersed the wax, freeing the acid.

**Example 4**

Sodium hypochlorite solution (13% active chlorine) at pH 12.5 was encapsulated under the following conditions by the biliquid column ejection method.

Pressure applied to pot	8 psig
Ejection velocity	5 m sec <sup>-1</sup>
Temperature of wall material	200°C
Temperature of core material	25°C
Immersion depth	1 mm
Orifice size	1 mm

Polyethylene wax MW 2000 and m.pt. 110°C was used as the wall material; the capsules obtained were approximately 2 mm in diameter and contained 50% core material. They were incorporated at the level of 25 per cm<sup>3</sup> in a household general purpose clear liquid cleaner of the following composition.

Sodium alkylbenzene sulphonate	%
	10
Kelzan gum*	0.4
Water	to 100

\*A xanthan gum supplied by ABM Industrial Products Limited

The composition was an effective general-purpose household cleaner containing about 0.3% active chlorine which could be released readily from the capsules by mechanical action during the cleaning process.

There was a loss of 50% of the active chlorine from this composition during storage at 25°C for 6 months. The same level of hypochlorite introduced directly into the liquid was, on the other hand, found to have disappeared completely after 3 weeks' storage under the same conditions.

**Example 5**

A general purpose household cleaning composition was prepared in the same manner as Example 4 except that the clear liquid cleaner had the following composition:

Sodium alkylbenzene sulphonate	%
	10
Benagel EW*	0.6
Water	to 100

\*A montmorillonite clay supplied by Berk Limited.

## Example 6

A clear fabric rinse conditioning liquid medium was prepared according to the following composition:

5	Hexadecyl trimethyl ammonium bromide	%	
		5	
	Sodium zylene sulphonate	3	
	Kelzan gum	0.5	
10	Water, perfume, etc.	to 100	

Droplets were prepared by first dispersing 12 parts of Photine C (RTM) a fluorescer of formula (4,4' - bis(2 anilino, 4 - diethanol-amino - 1,3,5triazine - 6 - yl amino) - 2,2' - (silbene disodium disulphonate) in 88 parts of paraffin wax (m.pt. 40°C); the slurry was then forced through a jet using the same operating conditions as for Example 2 except that a 0.6 mm jet was used in order to obtain droplets approximately 1 mm in diameter. These were incorporated into the liquid at a level of 16 per cm<sup>3</sup>.

The composition therefore contained about 0.1% fluorescer in a form which eliminated the well known inhibition of typical cotton-substantive fluorescers due to complex formation with the cationic active material. The effectiveness of the fluorescer in this compo-

sition was especially noticeable when it was used in a rinse following a wash process in which the action of the fluorescer normally present in the added wash product had been impaired; for example, when sodium hypochlorite solution, which will chemically degrade fluorescent materials in solution, had been added as a bleaching agent.

## Example 7

A shampoo containing encapsulated mineral oil as a general emollient and as an agent for improving the manageability of hair was prepared according to the following composition:

		% by weight	
	Clear Liquid medium	of liquid medium	
	Sodium lauryl triethoxy sulphate	15	45
	Nonyl phenol-12 ethylene oxide condensate	10	
	Ethyl alcohol	2	
	Laponite S synthetic clay (5% dispersion)	30	50
	Water, perfume	to 100	

Capsules were prepared according to Example 25 of US 3,423,489 under the following operating conditions:

55	Fill material	23°C	
	Temperature	0.25 mm	
	Fill orifice diameter	7.6 m/sec.	
	Fill ejection rate		
60	Shell material	70°C	
	Temperature	1.2 mm inner diameter	
	Shell orifice dimensions	1.6 mm outer diameter	
	Shell material purging velocity at orifice	0.5 m/sec.	

A 20% solution of the gelatine noted in Example 1 was used as the Shell material and liquid paraffin as the core. The mineral oil used to congeal the droplets was washed away with ice cold detergent solution and subsequently hardened with formalin as described in Example 1. The average diameter of the droplets was approximately 1 mm. and they contained about 60% core material. They were incorporated in the base liquid at the level of approximately 1000 per cm<sup>3</sup> therefore represent an embodiment of the invention in which a distinct visual appeal is provided by a relatively close packed array of droplets.

In addition it offers convenience in use as it is necessary to shake the product in order to obtain the correct proportioning of the medium and the capsules of the product.

As in the case of Example 1 the gelatin was readily dissolved above 40°C, thus releasing the contents of the capsules.

## WHAT WE CLAIM IS:—

1. A pourable liquid composition for cleaning or rinsing operations, which comprises a clear liquid medium and a visually distinct component, of particle size at least 0.5 mm diameter, stably dispersed and stably suspended in the medium. 85
2. A composition as claimed in claim 1 in which the component comprises 0.1—8.0% by weight of composition, and includes an agent which will provide a technical effect during use of the composition. 90
3. A composition as claimed in claim 2 in which the component comprises a protective inert carrier, around the agent. 95
4. A composition as claimed in claim 3 in which the carrier is a wax which breaks down during the use of the composition. 100
5. A composition as claimed in claim 3 in which the carrier is a gelatin which is tem-

perature responsive during use of the composition.

6. A composition as claimed in any preceding claim in which the component is of particle size between 1 mm and 1 cm diameter.

5 7. A composition as claimed in any preceding claim in which the medium has a turbidity of at most 5.

8. A composition as claimed in any preceding claim in which the component occupies not more than 10% by volume of the composition.

10 9. A composition as claimed in any preceding claim in which the component occupies 20—80% by volume of the total composition.

15 10. A composition as claimed in any preceding claim in which the medium has a yield value of not more than 2 dynes/cm<sup>2</sup>.

11. A composition as claimed in any preceding claim in which the medium comprises a suspending agent.

20 12. A composition as claimed in claim 11 in which the suspending agent is a swelling clay.

13. A composition as claimed in claim 11 in which the suspending agent is a soluble gum. 25

14. A composition as claimed in claim 11 in which the suspending agent is a montmorillonite. 30

15. A composition as claimed in any preceding claim in which the medium comprises an anionic synthetic detergent.

16. A composition as claimed in any preceding claim in which the medium is a non-ionic synthetic detergent. 35

17. A composition as claimed in any preceding claim in which the medium is coloured.

18. A composition as claimed in any preceding claim in which the component is coloured. 40

19. A composition substantially as described with reference to any one of Examples 1—7.

W. JAMIESON  
Chartered Patent Agent

Printed for Her Majesty's Stationery Office, by the Courier Press, Leamington Spa, 1973.  
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from  
which copies may be obtained.